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Radiation Effects in Candidate Materials for Spallation Neutron Environments — Program Summary for FY02

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Introduction

Candidate materials for the spallation neutron environment will require radiation tolerance to doses of 30 dpa at temperatures between 350 and 550¡C and injection of H and/or He in the range several hundred to a few thousand appm. [1] These are extremely severe and demanding conditions and basic data as well as understanding of material response to these conditions is lacking. Ultimately, experiments will need to be conducted in the actual environment in which the materials will function, but given the sparse database and understanding of radiation response under this these conditions, irradiation under approximate conditions will be extremely valuable.

Proton irradiation has proven to be an extremely effective tool in emulating the effects of neutron irradiation in austenitic stainless steel used in light water reactor cores. Radiation induced segregation, dislocation microstructure, irradiation hardening, void formation and susceptibility to irradiation assisted stress corrosion cracking are all properties that have been shown to be emulated by protons and with the proper dose dependence as well. [2] Proton irradiation of candidate spallation materials is planned with three objectives: 1) To generate some baseline irradiation effects data on promising alloys, 2) to begin to learn how these materials respond to irradiation and how the microstructure develops with dose, and 3) to assist in laying the foundation for future large scale irradiations, by identifying irradiation parameters and radiation effects mechanisms in candidate alloy systems of greatest potential importance.

Sample Fabrication

Samples were fabricated from both the HT-9 and T-91 alloys via electric discharge machining at Shular Tool Co, in Oak Ridge, TN. A schematic of the TEM sample design is illustrated below in Figure 1. Ten samples of each alloy were made, allowing for several spares from each alloy. Six specimens were needed from each alloy for irradiations to 3, 7, and 10 dpa, with and without He at each dose. Up to three 3-mm TEM discs can be cut from each bar. Up to eight samples of this size can be irradiated at once, allowing for flexibility in proton irradiations.

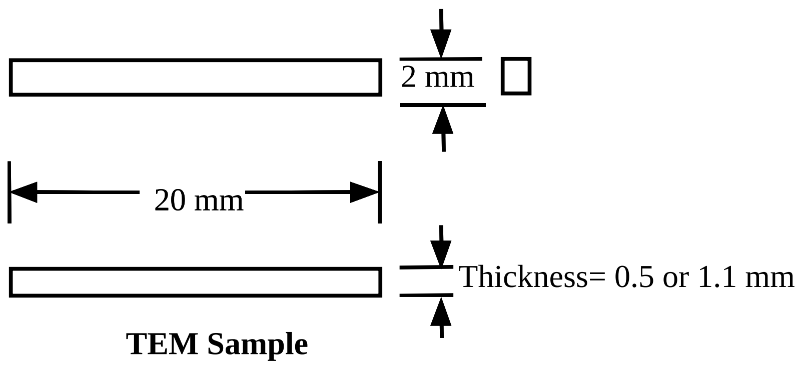

Figure 1: Schematic of TEM samples fabricated from HT-9 and T-91 alloys.

Helium Implantations

Prior to irradiation, five samples (three for irradiation and two spares) from each alloy were implanted with 2.3 MeV He at room temperature. Helium implantation was performed using the Tandetron Accelerator in the Michigan Ion Beam Laboratory at the University of Michigan. A uniform He distribution was achieved by controlling both the angle of incidence between the samples and He beam and the number of He particles implanted at any given angle. The samples were rotated from 0 to 70° in 5° increments. The number of He ions incident on the samples for each angle was also monitored using a beam current integrator. The resulting simulated He profile and He content at each angle are illustrated in Figure 2. The He concentration is uniform at ~ 100 appm between 1 and 4 μm. The He implantation was performed in four 25 appm steps, each taking approximately 5 hours to complete.

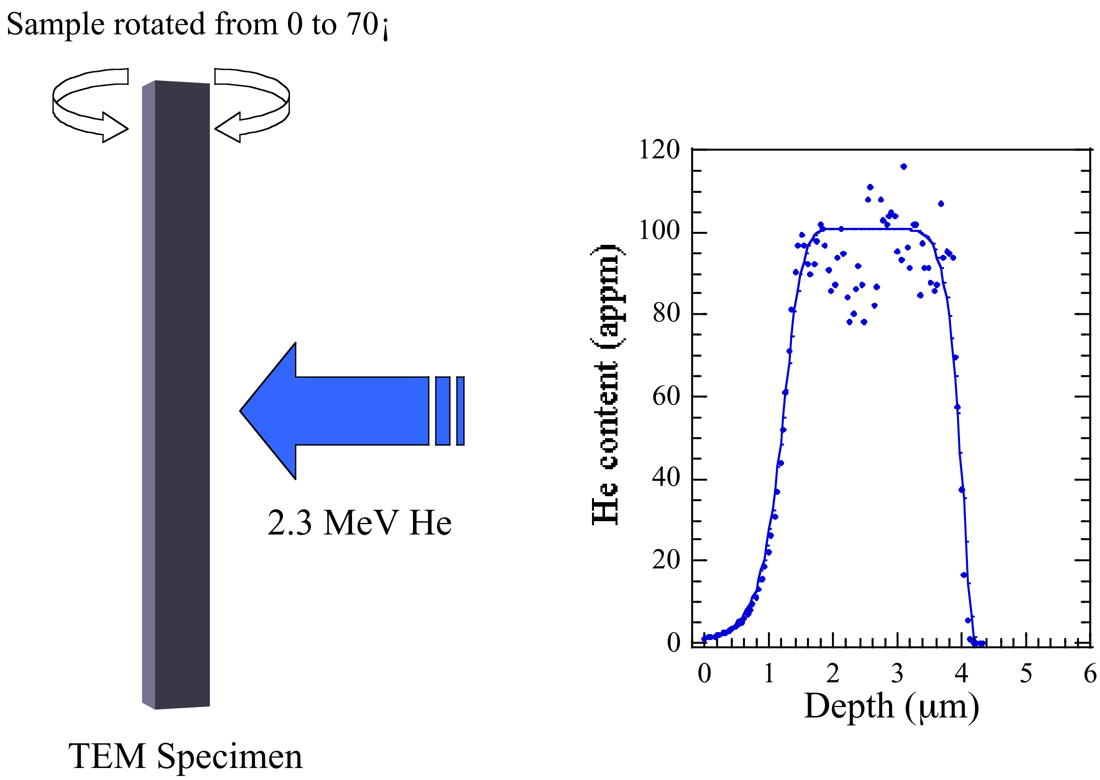

Figure 2: Schematic of He implantations into TEM specimen and resulting He concentration as a function of depth.

Proton Irradiations

Proton irradiations were performed using a specially designed stage connected to the General Ionex Tandetron accelerator at the Michigan Ion Beam Laboratory. Irradiations were conducted using 2.0 MeV protons at a dose rate of approximately 2 x 10^{-5} dpa/s (the experimental doses and dose rates are calculated using the SRIM2000 simulation), resulting in a nearly uniform damage rate through the first 15 μm of the proton range (20 μm), where dpa is calculated using SRIM with a displacement energy of 40 eV. The calculated dose rate is plotted as a function of depth for the HT-9 and T-91 alloys in Figure 3. Also illustrated in Figure 3 is the He distribution. Irradiations were conducted to 3, 7, and 10 dpa at 450°C.

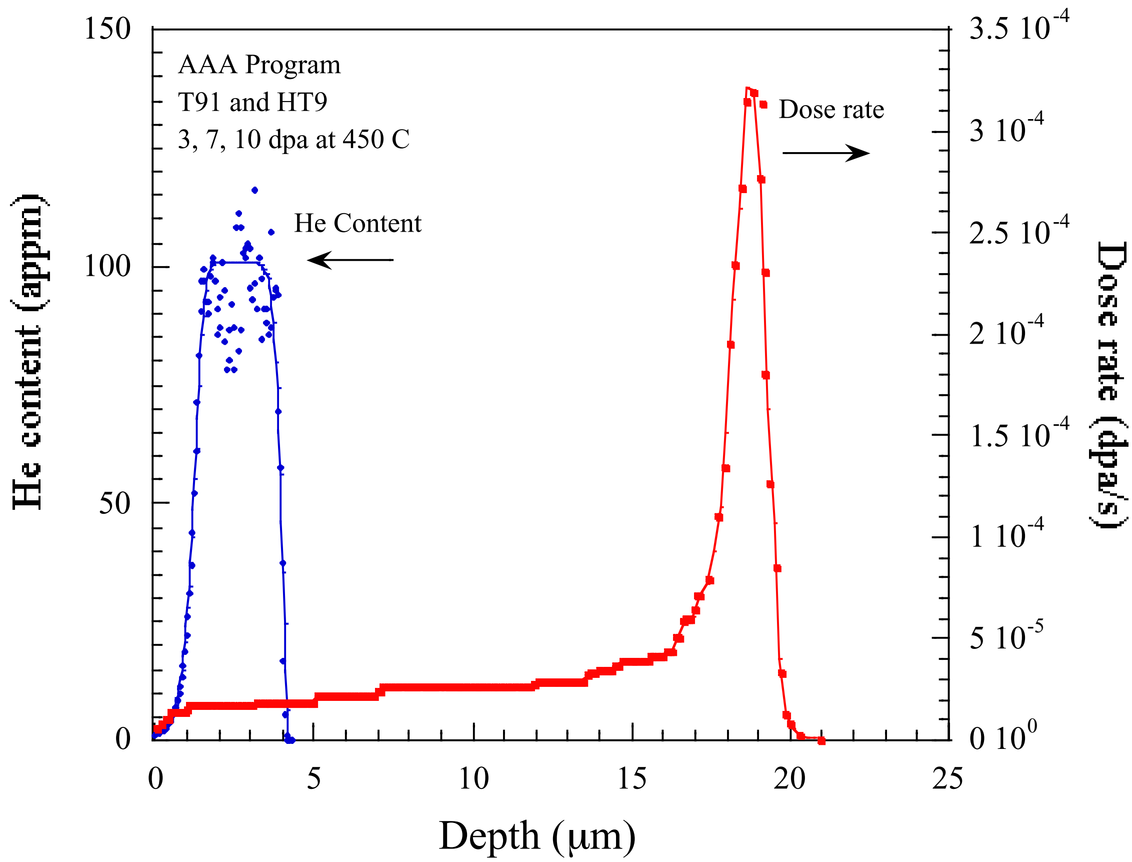

Figure 3: Dose rate and He concentration as a function of depth for HT-9 and T-91 alloys.

The irradiation stage was designed to control the sample temperature by controlling the stage temperature. The stage was heated using a resistive cartridge heater and cooled using room temperature air flowing through cooling lines that penetrated the back of the stage. The stage surface is made of copper to provide good heat conduction away from the samples. To provide effective thermal contact between the sample bars and the stage, a thin layer of indium was placed between the samples and the stage surface. Indium melts at 156°C and is molten at the irradiation temperature, maximizing the thermal contact between samples and stage. Sample temperature was monitored using two techniques. Type J (iron/constantan) thermocouples were spot welded directly to sample surfaces to provide one temperature measurement. Typically, three to five thermocouples were attached to the samples during any one irradiation. A separate thermocouple monitored the temperature at the back of the stage. In addition to the thermocouples, a calibrated infrared pyrometer monitored the surface temperature of the samples during irradiation. The pyrometer was also controlled remotely to scan the surface of the specimens to insure a uniform temperature. The pyrometer was calibrated prior to irradiation by heating the samples with the cartridge heater to the set-point temperature and adjusting the pyrometer s emissivity setting so that the pyrometer reading matched that of the thermocouples. During irradiation, the sample temperature was controlled to – 10¡C of the set point temperature (450 °C) by controlling the amount of heating and/or cooling provided to the stage. By providing a large fraction of the total heat input to the samples from the cartridge heater, temperature fluctuations due to fluctuations in beam current were minimized.

The irradiation stage was electrically isolated from the beam line and four rectangular tantalum apertures were used to define the area on the sample bars that was irradiated with the proton beam. The approximately 3 mm diameter proton beam was raster-scanned across the stage so that about half the total beam current was deposited on the samples and half on the apertures. Raster-scanning ensured that samples at any position on the stage received the same dose. Additionally, balancing the amount of current on each of the apertures centered the proton beam.

Experimental parameters were tracked continuously during irradiation using a PC-based monitoring system. The monitoring software recorded the stage current, current for each of the apertures, pyrometer temperature and up to five thermocouple temperatures. This system allowed the operator to continuously monitor experimental parameters while also providing a comprehensive history of each irradiation. Alarms were installed to alert the operator when experimental parameters moved outside acceptable limits.

The first proton irradiation consisted of four samples each of the T-91 and HT-9 alloys (two of each alloy with He and two without He). The first irradiation was conducted to a dose of 3.0 dpa. The beam current and temperature history for this irradiation is illustrated in Figure 4. Beam current was extremely stable during the irradiation, with up to 75 uA on the samples at times. This is the highest beam current ever used during a proton irradiation in this laboratory. Sample temperature was also very stable during the 58-hour irradiation, as illustrated in Figure 4. Temperature profile was also monitored regularly by moving the pyrometer over the surface of the samples. A sample temperature profile is illustrated in Figure 5. All temperatures are within 5°C of the target temperature of 450°C.

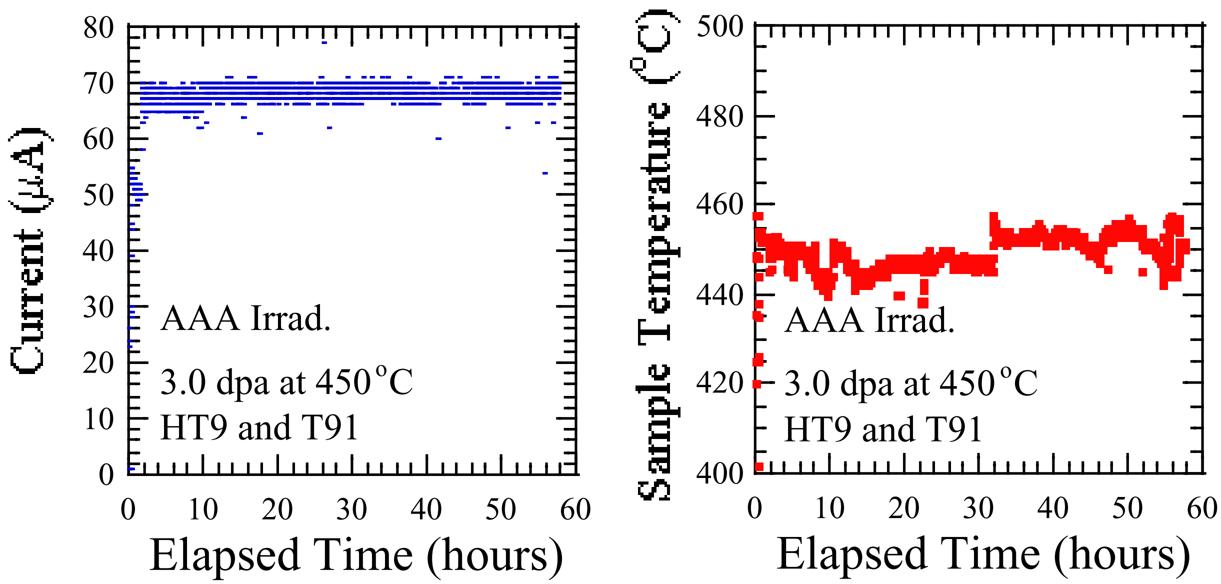

Figure 4: Sample temperature and proton beam current on samples during proton irradiation of T-91 and HT-9 to 3.0 dpa at 450°C.

Samples for the second proton irradiation consisted of two unirradiated samples each of the T-91 and HT-9 alloys (one of each alloy with He and one without He). Two samples of each alloy irradiated to 3.0 dpa (one of each alloy with and without He) in the first irradiation were also included in the second irradiation. After an additional 7.0 dpa, the total dose on these four specimens was 10.0 dpa. The beam current and temperature history for this irradiation is illustrated in Figure 6. Beam current was extremely stable during the second irradiation as well, again with up to 75 uA on the samples at times. There was no interruption of beam during the entire 126-hour irradiation. Sample temperature was again very stable as illustrated in Figure 7. A sample temperature profile is illustrated in Figure 8. All temperatures are within 5°C of the target temperature of 450°C.

Proton irradiation also induces a very low level of radioactivity; the result of proton-capture reactions with Fe. Interactions with vanadium are another possibility, but must be confirmed. The majority of the residual radioactivity was in the form of β-decay and was measured using a gas proportional detector. The samples were each counted for five minutes in a Tennelec LB 5100 2π alpha/beta counter. The activity of each sample was normalized to the sample area and compared the activity of other specimens from the irradiation batch to determine the dose uniformity. The counting results for all irradiations are shown in Table 1. Counting was uniform between samples of the same alloy at the same dose level.

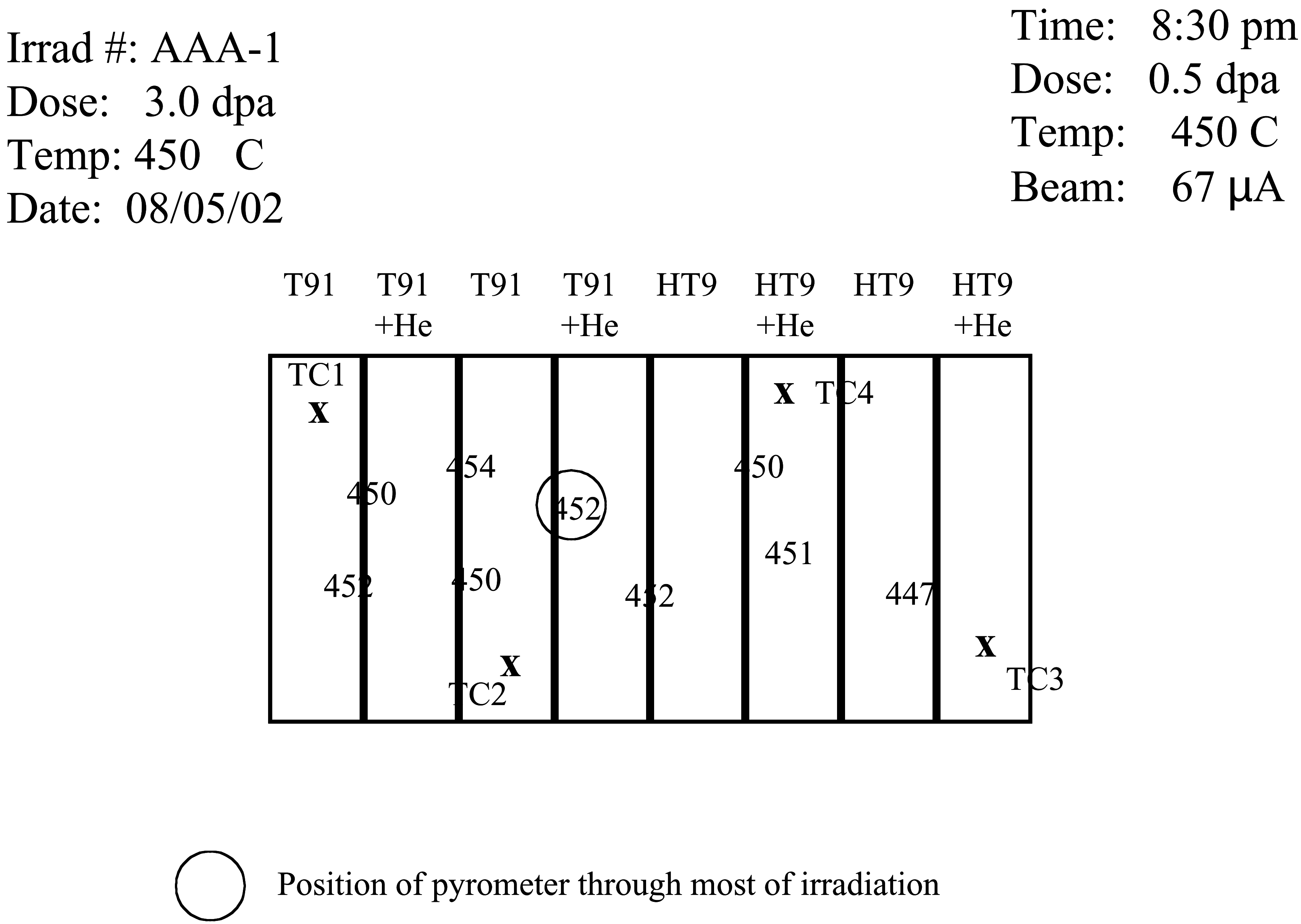

Figure 5: Sample temperature profile taken during 3.0 dpa irradiation of T-91 and HT-9 samples at 450°C. All measured temperatures are within 4°C of the target temperature.

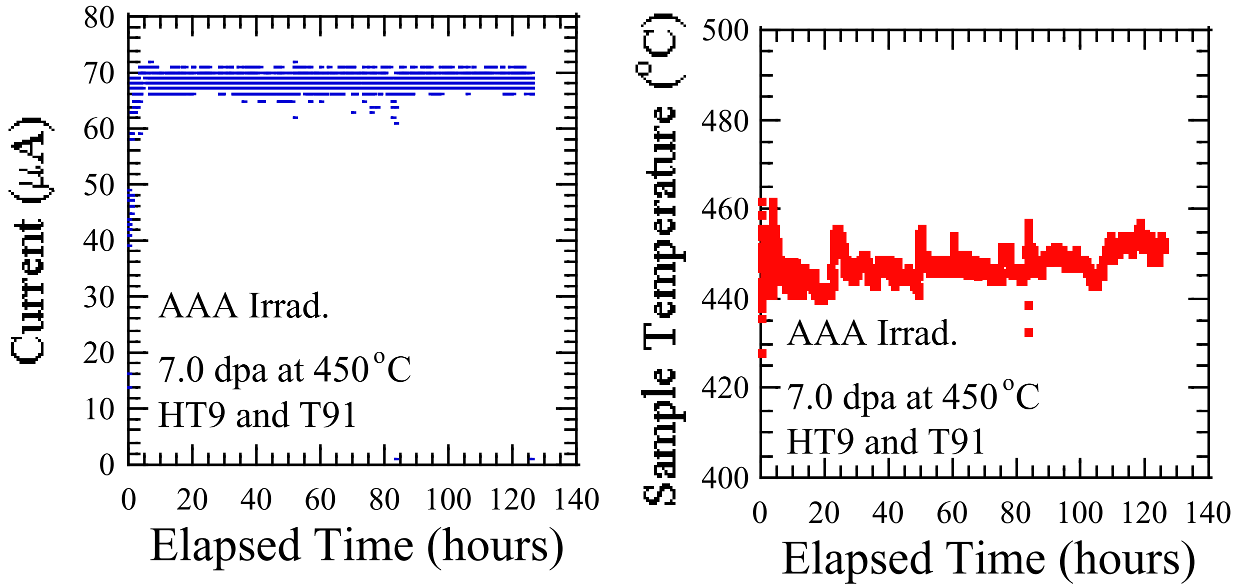

Figure 6: Sample temperature and proton beam current on samples during second proton irradiation of T-91 and HT-9 to 3.0 dpa at 450°C.

Table 1: Activity counting results for proton-irradiated samples of T-91 and HT-9 alloys.

	Counts (#/mm^2/dpa)	
Alloy/dose	No He	100 appm He
T91: 3.0 dpa	140	112
T91: 7.0 dpa	87	96
T91: 10.0 dpa	281	296
HT9: 3.0 dpa	195	179
HT9: 7.0 dpa	158	154
HT9: 10.0 dpa	290	272

Following measurement of activity, shipment of the low-level radioactive HT-9 samples to ANL was arranged through the University of Michigan Health Physics Department. Samples were delivered to Argonne-National Laboratory-West for TEM analysis on September 3, 2002. Irradiated T-91 samples will be shipped to Los Alamos National Laboratory for analysis of the irradiated microstructure.

References

1. Journal of Nuclear Materials, Volume 301, No.1, 2002.
2. G. S. Was, J. T. Busby, T. Allen, E. A. Kenik, A. Jenssen, S. M. Bruemmer, J. Gan, A. D. Edwards, P. Scott and P. L. Andresen, "Emulation of Neutron Irradiation Effects with Protons: Validation of Principle," J. Nucl. Mater., 300 (2002) 198-216.

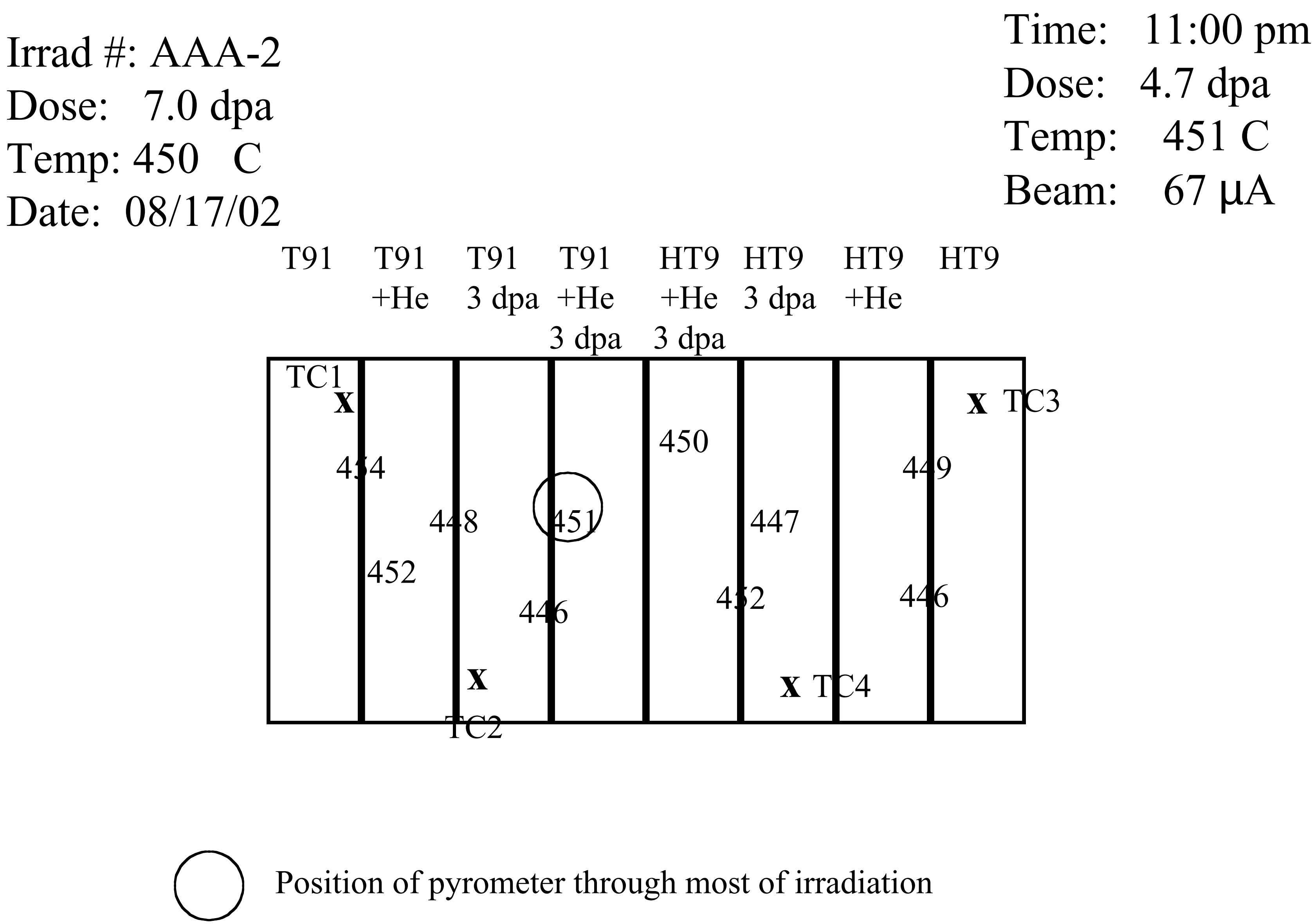

Figure 7: Sample temperature profile taken during 7.0 dpa irradiation of T-91 and HT-9 samples at 450°C. All measured temperatures are within 4°C of the target temperature.